

# TOPOLOGICAL CORRELATIONS IN SOAP FROTHS

K.Y. Szeto, T. Aste\* and W. Y. Tam

*Department of Physics, The Hong Kong University of Science and Technology, phszeto@usthk.ust.hk  
Clear Water Bay, Kowloon, Hong Kong.*

*\*Equipe de Physique Statistique, LUDFC Université Louis Pasteur,  
3, rue de l'Université, 67084 Strasbourg France. tomaso@ldfc.u-strasbg.fr  
(February 1, 2008)*

Correlation in two-dimensional soap froth is analysed with an effective potential for the first time. Cells with equal number of sides repel (with linear correlation) while cells with different number of sides attract (with NON-bilinear) for nearest neighbours, which cannot be explained by the maximum entropy argument. Also, the analysis indicates that froth is correlated up to the third shell neighbours at least, contradicting the conventional ideas that froth is not strongly correlated.

PACS numbers:82.70Rr, 02.50.-r, 05.07.Ln

Two-dimensional cellular structures are space-filling disordered partitions of space by cells that are irregular polygons and often appear in nature as in metal grains, biological tissues and common soap froths [1]. These cellular patterns are usually trivalent (3 edges meet at a vertex) due to topological stability. A generic term for these networks is “froth”, since the soap-froth is the archetype of such structures. Many theoretical studies on froth are based on mean field, with minimum consideration on the correlation between cells [2,3]. This is understandable as the conventional wisdom assumes small correlation effect in froth, which is almost completely described by the Aboav-Weaire law [4–6]. Also, any theoretical description taken correlation effects into account is much more difficult. Thus, most of the approaches on froths make some kind of independent bubble approximation, and neighbours are described by a mean field [2] or as random neighbours [3]. It is our aim to test these approaches by analysing the experimental data on real dry soap froth and by extracting the topological correlation function between cells. We find that there are very strong and long range correlation, implying independent bubbles approaches are unrealistic. The data indicate that any reasonable description respecting the correlation effects on froth must go at least to the third shell neighbours. Furthermore, these correlations effects are not at all well described by the maximum entropy arguments [7], as the predicted bilinearity for the correlators is only a rather crude approximation to the data. The data therefore strongly suggest that there must be a new theory describing real soap froth, with full consideration of the energies and the correlations.

In froths a cell can be characterized topologically by its number of sides. Froths are disordered systems and a topological, statistical description of their structure is given by the correlation function. To extract the relevant structural properties from a disordered system like a froth is a very difficult task. Indeed, the absence of symmetries and periodicities requires a priori the knowledge of the information about all the cells in the system. A simple and useful approach is to analyze the froth as structured in concentric layers of cells at the same topological distance  $j$  from a given central cell [8–11] (where the topological distance between two cells is the minimum number of edges that a path must cross to go from one cell to the other [9–11]). An interesting and meaningful quantity in this analysis by concentric layers is, for instance, the number of cells in a given layer ( $K_j(n)$ , with  $n$  the number of sides of the central cell) [10]. Here we focus on the correlation function. The two-cell topological correlation function ( $C_j(n, m)$ ) is the probability of finding a cell with  $m$  sides at a distance  $j$  from a cell with  $n$  sides. The two-cell topological correlation function is equal to the total number  $N_j(n, m)$  of couples of cells with respectively  $n$  and  $m$  sides which are at a relative distance  $j$ , divided by the number of all couples of cells at a relative distance  $j$

$$C_j(n, m) = \frac{N_j(n, m)}{\sum_{n, m=3}^{\infty} N_j(n, m)} \quad . \quad (1)$$

We consider the  $n$ - and  $m$ - sided cells as distinguishable. Therefore each couple is counted twice and  $N_j(n, m)$  and  $C_j(n, m)$  are symmetric in  $n, m$  (i.e.  $C_j(n, m) = C_j(m, n)$ ).

By definition, in uncorrelated systems the correlation function must factorize:  $C_j(n, m) = s_j(n)s_j(m)$ . The normalization and the symmetry of  $C_j(n, m)$  imply  $\sum_m s_j(m) = 1$ , consequently  $s_j(n) = \sum_m C_j(n, m)$ . Now, we calculate this quantity and we interpret it in term of the properties of the layers. Consider two cells with  $n$  and  $m$  sides which are at a relative topological distance  $j$ . Such a pair, connected by a path of length  $j$ , can be seen as a “string” with  $n$ - and  $m$ -sided terminations. Clearly the number of these strings in the cellular system is  $N_j(n, m)$  (where each string is counted twice since the terminations are distinguishable cells). The number of strings of length  $j$  with a termination in a cell with  $n$  sides and the other termination free is given by  $\sum_m N_j(n, m)$ . The same quantity is equal to the number of  $n$ -sided cells in the system ( $N(n)$ ) multiplied by the average number of strings of length  $j$  which terminate in one  $n$ -sided cell ( $K_j(n)$ ). Therefore,  $\sum_{m=3}^{\infty} N_j(n, m) = N(n)K_j(n)$ . Finally, the total number of strings of length  $j$  is equal to the total number of cells ( $N_T$ ) multiplied by the average number of strings of length  $j$  which terminate in any given cell ( $\langle K_j \rangle$ ), giving

$$\sum_{n, m=3}^{\infty} N_j(n, m) = \sum_{n=3}^{\infty} N(n)K_j(n) = N_T \sum_{n=3}^{\infty} \frac{N(n)}{N_T} K_j(n) = N_T \langle K_j \rangle \quad , \quad (2)$$

where  $\langle \dots \rangle = \sum_n p(n) \dots$ , with  $p(n) = N(n)/N_T$  being the probability of an  $n$ -sided cell in the whole froth. The probability to find a string of length  $j$  with a termination in a cell with  $n$  sides is consequently given by the ratio

$$s_j(n) = \frac{\sum_{m=3}^{\infty} N_j(n, m)}{\sum_{n, m=3}^{\infty} N_j(n, m)} = \sum_{m=3}^{\infty} C_j(n, m) = \frac{N(n)K_j(n)}{N_T \langle K_j \rangle} = p(n) \frac{K_j(n)}{\langle K_j \rangle} \quad . \quad (3)$$

By following this point of view, the correlation function  $C_j(n, m)$  can be interpreted as the probability of having a string of length  $j$  with terminations in two cells with  $n$  and  $m$  sides respectively. In an uncorrelated system this conditional probability must be the product of the two string probabilities

$$C_j^{un}(n, m) = s_j(n)s_j(m) = \frac{K_j(n)K_j(m)}{\langle K_j \rangle^2} p(n)p(m) \quad . \quad (4)$$

Even when the system is uncorrelated, taken two cells at distant  $j$ , the probability to have one cell with  $m$  sides and the other cell with  $n$  sides is not given by the simple product of the probabilities of finding independently an  $n$ - and an  $m$ -sided cells ( $p(n)p(m)$ ). The factor  $K_j(n)K_j(m)/\langle K_j \rangle^2$  in Eq.(4) indicates that a cell in a froth (even in an uncorrelated one) CANNOT be topologically independent by its neighbours. Indeed, in froths, a single isolated cell doesn't exist. The cell, its number of sides, and the number of neighbours at any distance  $j$  constitute a unique system. (Note that, for  $j = 1$ , the function  $\frac{C_1(n, m)}{C_1^{un}(n, m)} - 1 = \beta_{n, m}$  is the topological short-range order coefficient introduced by Le Caër et al. [12].)

The structure of a froth can be studied in term of effective “potential” between different cells. These potentials can correspond to a real dynamical interactions between coupled cells during the process of formation and evolution of the system; or they can simply indicate the degrees of affinities of two cells to stay at a given relative distance. Attractive interactions are associated to couples of cells which appear in the froth with a higher probability than in the uncorrelated case, while

negative interactions correspond to the opposite case. Without any loss in generality one can define the correlation function of the form

$$C_j(n, m) = C_j^{un}(n, m) \exp \left( -\beta \varphi_j(n, m) \right) . \quad (5)$$

Here  $\varphi_j(n, m)$  is the interaction effective potential between two cells with  $n$  and  $m$  sides at topological distance  $j$ , and  $\beta$  is the inverse temperature. This potential is zero in uncorrelated systems, negative when two cells attract and positive when they repel.

We have analyzed data for soap froths prepared at different times, all in the scaling regime [13,14]. In Fig.1, we show the variation of  $C_j(n, m)/C_j^{un}(n, m)$  (Fig.1a) and  $\varphi_j(n, m)$  (Fig.1b) vs topological distance  $j$ . These correlations and potentials are the same for a sample that is 6 hours in the scaling regime as another sample that is 14 hours in the scaling regime. Other times have also been checked and  $C_j(n, m)/C_j^{un}(n, m)$  and  $\varphi_j(n, m)$  are time independent within experimental errors. From Fig.1a, it is clear that soap froth is strongly correlated at least up to the third neighbours. Cells with equal number of sides (e.g.  $(n, m) = (5, 5), (6, 6), (7, 7)$ ) have lower probability to be first neighbours than in the uncorrelated case and the contrary is for cells with different number of sides (see Fig.1a for  $j = 1$ ). The associated effective potential between first neighbours is therefore repulsive for equal sided cells and attractive for cells with different number of sides (see Fig.1b). This is the Aboav law [4] in the language of correlation. Increasing the topological distance  $j$  the correlation and the potential shows oscillations which can be associated to the screening and anti-screening of the effective interaction.

In accordance with previous analysis on first neighbours correlation [12,15,16], we introduce the correlator  $A_j(n, m) \equiv [C_j(n, m)/p(n)p(m)]$ , which is the probability to have an  $n$ -sided cell at a distance  $j$  from an  $m$ -sided cell, given that the two cells exist (this correlator become approximately equal to the Boltzmann factor  $e^{-\varphi_j(n, m)}$  as  $s_j(n) \rightarrow p(n)$  for  $j \rightarrow \infty$ ). Fig.2 shows the plot of  $A_j(n, m)$  vs  $m$  for fixed  $n = 4, \dots, 8, n \neq m$  and fixed  $j = 1, 2, 3$ . There are theories [7,17] that predict the bilinearity of  $A_1(n, m)$  in  $n, m$  using maximum entropy argument. The poor fit to straight line of the data in Fig.2, and the strong deviation when  $n = m$ , indicate that the bilinearity of  $A_j(n, m)$  is at best a very crude approximation. Afterall, this disagreement should not be too surprising as the potential, as defined in Eq.5, manifests strong and long range interaction present in soap froth. On the other hand, we find a surprisingly simple linear behavior of  $A_j(n, n)$  vs  $n$  for  $j = 1, 2, 3$ . Fig.3 shows the plot of  $A_j(n, n)$  vs  $n$  for fixed  $j = 1, 2, 3$ . The data can be described by the linear function  $A_j(n, n) = \mu(j)n + \lambda(j)$  with coefficients  $\mu(1) = 0.193 \pm 0.008, \lambda(1) = -0.60 \pm 0.05$ ;  $\mu(2) = 0.121 \pm 0.005, \lambda(2) = 0.36 \pm 0.03$  and  $\mu(3) = 0.073 \pm 0.003, \lambda(3) = 0.69 \pm 0.02$ .

In conclusion, the data we present on soap froth strongly suggest that correlation effects are of paramount importance in our understanding of the quasi-static and dynamics of froth. The data indicate that the correlation is important up to the third shell at least, with linear correlator for cells of same number of edges, but non-bilinear behaviour for cells of different number of edges. Future theories of froth dynamics should include these strong correlations.

## Acknowledgements

K.Y. Szeto acknowledges support from the Hong Kong Telecom Institute of Information Technology. T. Aste acknowledges discussion with N. Rivier and D. Dubertret and the support from the Hong Kong University of Science and Technology and the partial support from EU, HCM Program "Physics of Foams" CHRXCT940542 and by the TMR contract ERBFMBICT950380. W.Y. Tam acknowledges support from the Direct Allocation Grant 96/97.SC22 of the Hong Kong University of Science and Technology.

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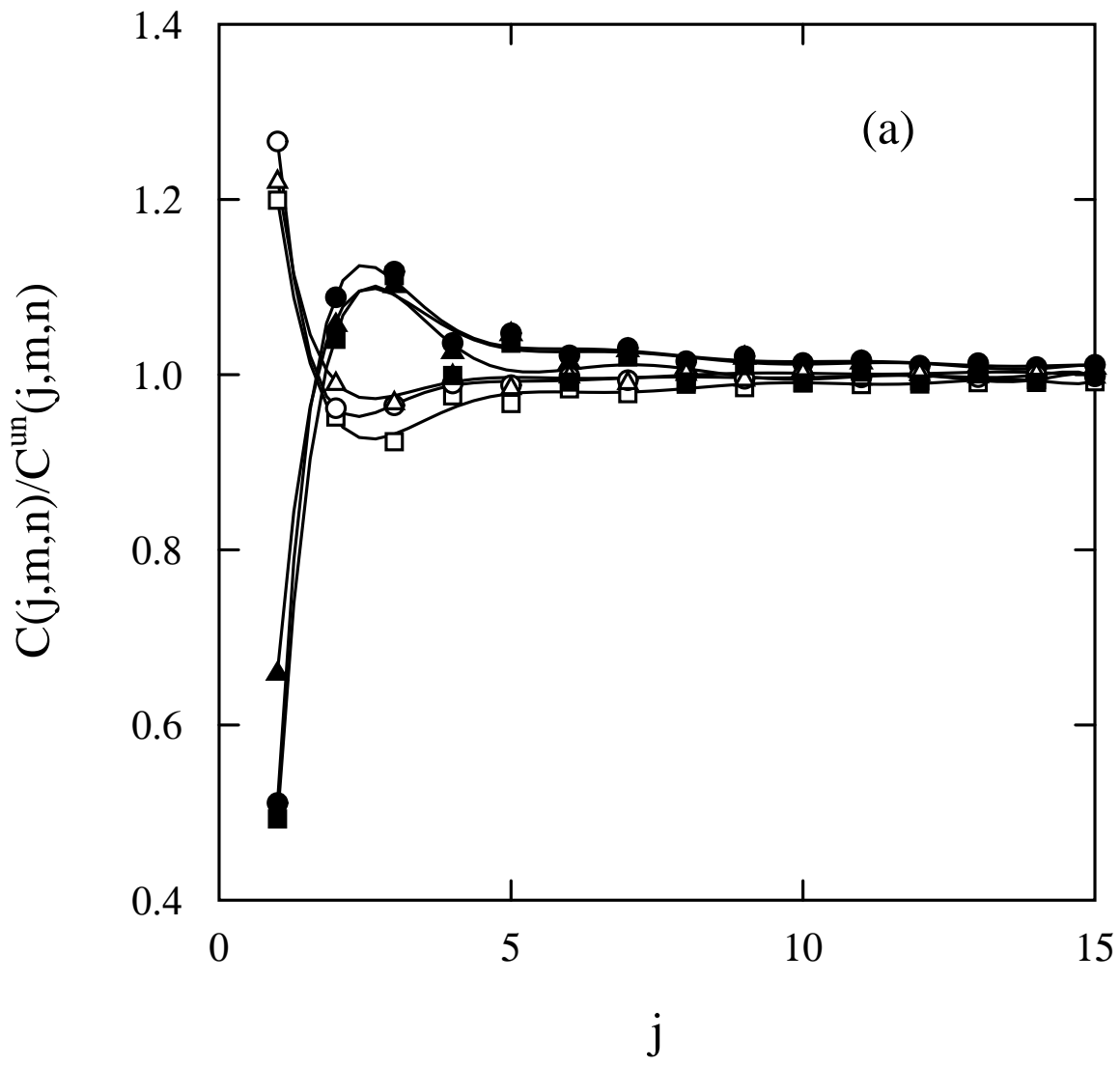
## Figures

Fig.1 (a) Relative correlation  $C_j(n, m)/C_j^{un}(n, m)$  and Fig.1 (b) Effective potential  $\varphi_j(n, m)$  vs topological distance  $j$  for couples of soap froth cells with  $n$  and  $m$  sides, and  $(n, m) = (5, 5)$ , solid circle,  $(n, m) = (6, 5)$ , open square,  $(n, m) = (6, 6)$ , solid triangle,  $(n, m) = (7, 5)$ , open circle,  $(n, m) = (7, 6)$ , open triangle,  $(n, m) = (7, 7)$ , solid square

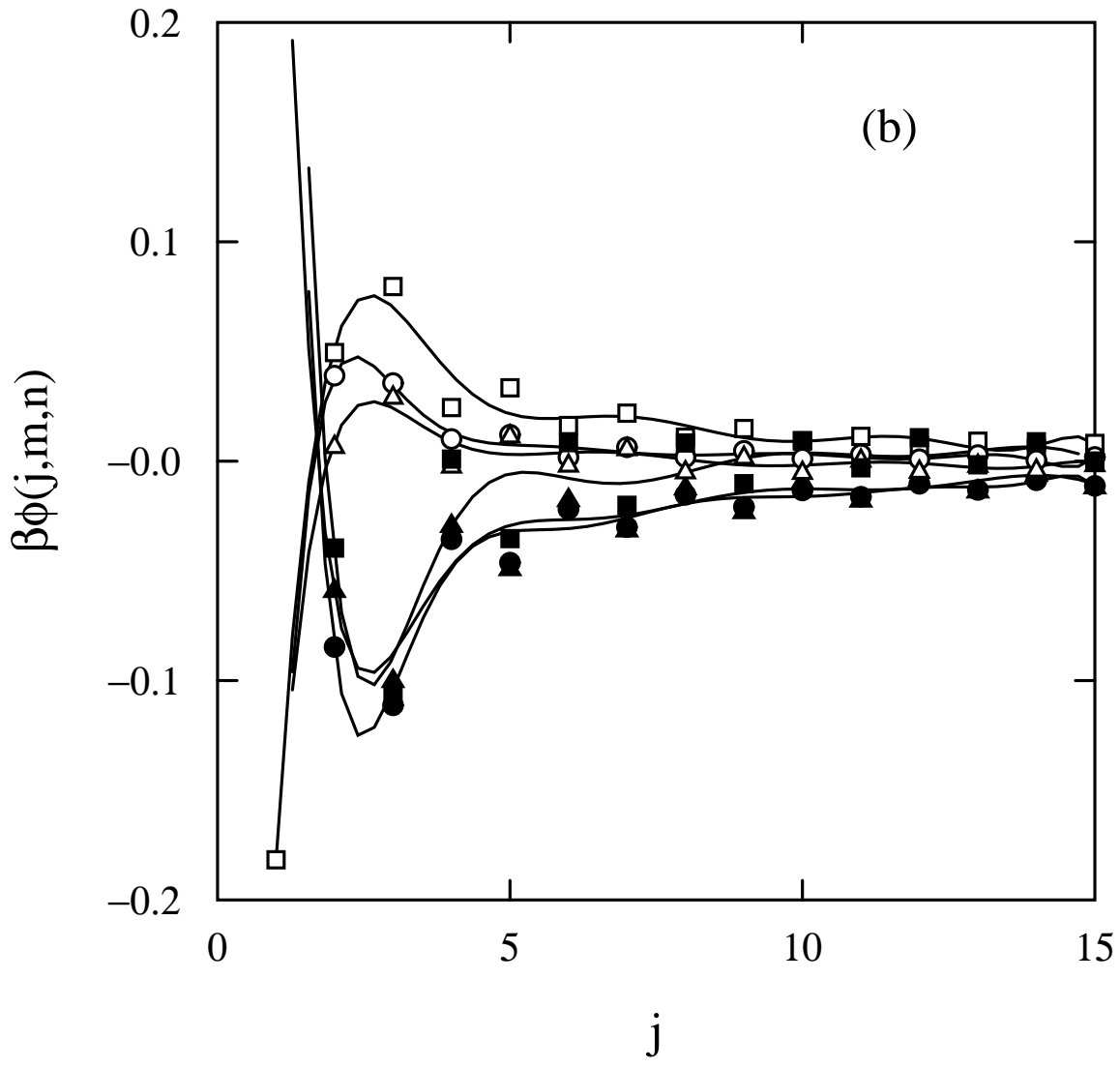
Fig.2 Correlator  $A_j(n, m)$  vs  $m$  for fixed  $n \neq m$ , and (a)  $j = 1$ , (b)  $j = 2$ , and (c)  $j = 3$ . Here  $n = 4$ , open circle,  $n = 5$ , solid circle,  $n = 6$ , open triangle,  $n = 7$ , solid triangle,  $n = 8$ , open square

Fig.3 Correlator  $A_j(n, n)$  vs  $n$  for fixed  $j$ ; (a)  $j = 1$ , open circle, (b)  $j = 2$ , open triangle, (c)  $j = 3$ , open square

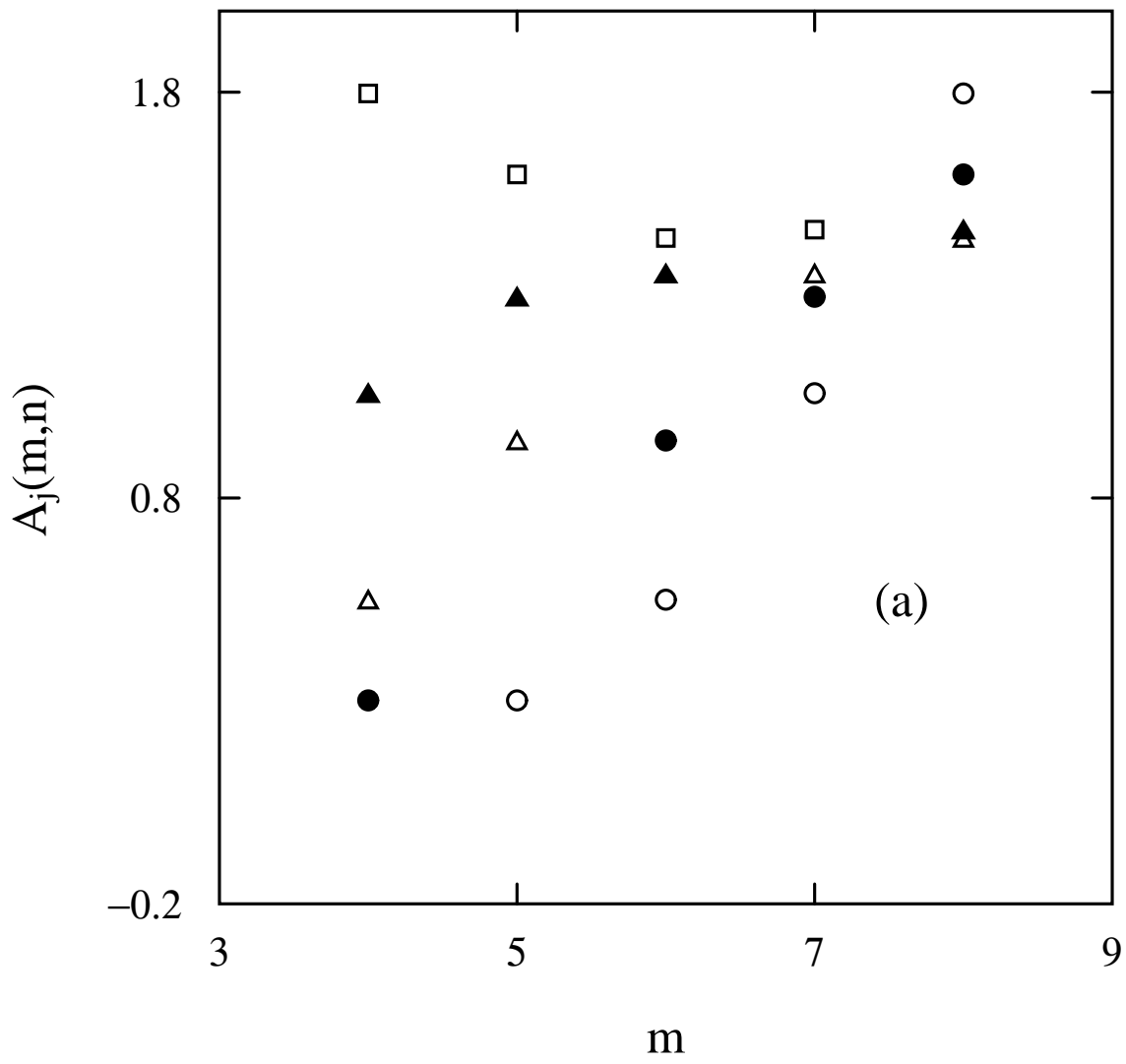
Szeto et al Fig. 1a

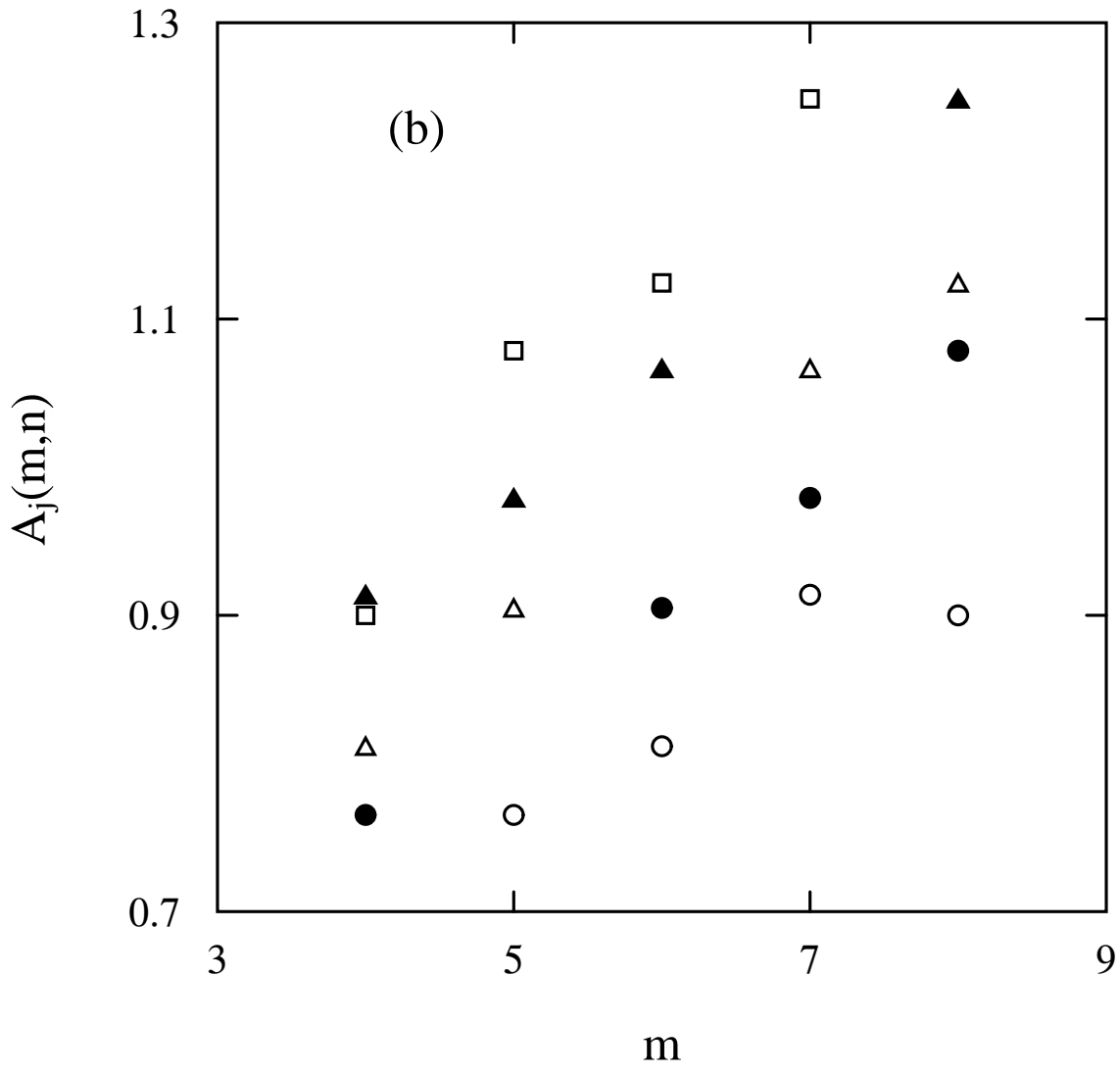


Szeto et al Fig. 1b

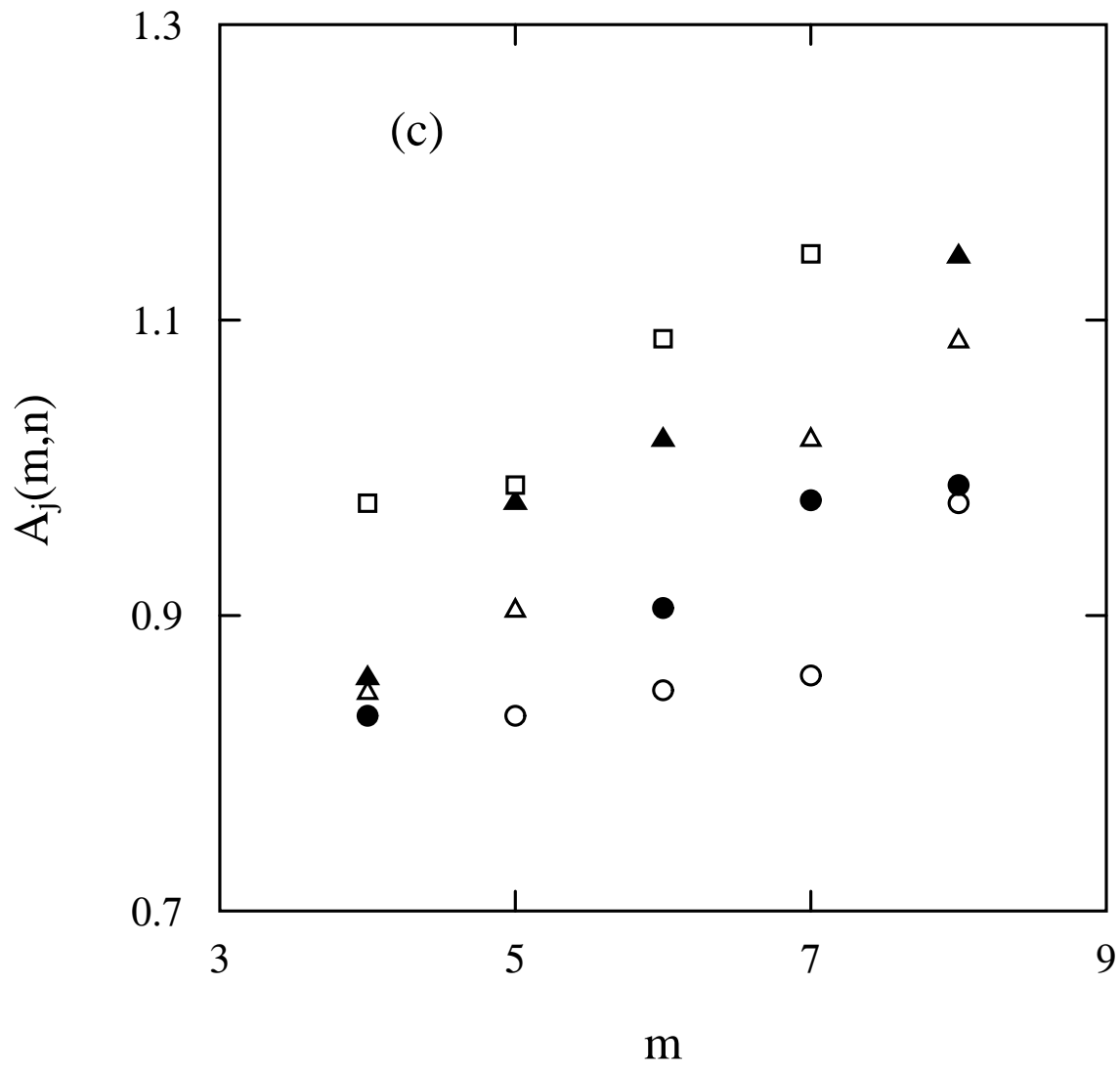


Szeto et al Fig. 2a









Szeto et al Fig. 3

